Strain-induced coarsening in nanocrystalline metals under cyclic deformation

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Abstract.

Atomic-scale computer simulations have previously identified a deformation mechanism, which becomes important in nanocrystalline metals with grain sizes below 10—15 nm. Instead of proceeding through dislocation activity in the grains, the deformation occurs by slip events in the grain boundaries, leading to a reverse Hall-Petch effect, i.e. a decrease in hardness with decreasing grain size. In this paper, the consequences of this shift in deformation mode are investigated for systems subjected to large strains in a cyclic deformation pattern.

In most coarse-grained metals, severe plastic deformation leads to grain refinement. Indeed, severe plastic deformation is often used to generate nanocrystalline metals with grain sizes down to hundred nanometres. The simulations indicate that these processes are suppressed in sufficiently small grains, and instead the sliding events in the grain boundaries dramatically enhance diffusion processes, and lead to grain coarsening as the deformation proceeds.

1. Introduction.

One of the attractive properties of nanocrystalline metals is their much-increased hardness compared to coarse-grained metals. The increase in hardness is caused by the Hall-Petch effect [1,2], the hindering of the motion (and perhaps creation) of dislocations by grain boundaries. There has been much controversy over the behaviour at the smallest grain sizes, starting with the observations by Chokshi *et al.* of a reverse Hall-Petch effect for the smallest grain sizes [3]. In particular, it is not clear if the observed reverse Hall-Petch effect is caused by reduced sample quality at the smallest grain sizes, or if it is indeed an intrinsic effect (for a discussion, see e.g. Morris [4]). Computer simulations are one way to shed light on the behaviour of nanocrystalline metals, in particular for the smallest grain sizes, where experimental investigations are the most difficult. Computer simulations of nanocrystalline copper, palladium and nickel have shown that a different deformation mechanism is active in nanocrystalline metals with grain sizes below 10—15 nm [5,6,7,8,9,10,11]. At these small grain sizes, the large density of grain boundaries essentially prevents dislocation motion from being an efficient mechanism to relieve the stress. Instead, the metal deforms by grain boundary sliding. This is possible as a significant fraction of the atoms is in the grain boundaries. In this regime, decreasing the grain size leads to a decreased hardness. It is thus clear that there must be a maximum in hardness at an optimal grain size [12,13].

Until now, molecular dynamics simulations of plastic deformation in nanocrystalline metals have focused on the early stages of plastic deformation (up to around 10%

strain.) In this work, the behaviour at larger strains is investigated, with the purpose of investigating, how the grain-boundary mediated deformation mechanism influences the behaviour at large plastic strains.

2. Simulation methods.

Samples of nanocrystalline copper are generated using a Voronoi construction: a set of grain centres are chosen at random, and the part of space closer to a given centre than to any other centre is filled with atoms in a face-centred cubic lattice with a randomly selected crystal orientation. Periodic boundary conditions are applied in all directions to mimic that the system is deep within the bulk of a larger sample. The system studied here contains 100000 atoms in 16 grains, corresponding to a system size of $(10.6 \text{ nm})^3$ and a grain diameter of 5.5 nm. To allow unfavourable configurations in the grain boundaries to relax, the system is briefly annealed by running a 50 ps molecular dynamics simulation at 300 K. For further details, please see ref. [6].

To evolve the system in time, a constant-temperature molecular dynamics simulation is performed. The interatomic forces are described within the Effective Medium Theory (EMT) [14,15]. To keep the temperature approximately constant, Langevin dynamics [16] is used: a small friction and a fluctuating force are added to the equation of motion of the atoms. While the simulation is running, the sample is deformed at a constant strain rate by changing one of the dimensions of the unit cell, while adjusting the two other ones such that the transversal components of the stress remain zero. The time step of the simulation is 5 femtoseconds. The software used for these simulations is freely available on the Internet as part of the CAMP Open Software project [17].

A cyclic deformation pattern was chosen. The system is deformed under tension until a nominal strain of 10% is reached, then the deformation direction is reversed and the sample is compressed until a strain of -10% is reached, the deformation is then reversed again and the sample is elongated back to the initial dimensions (0% strain). These cycles are repeated multiple times.

3. Results.

Figure 1 shows a sample of nanocrystalline copper after 0, ½, 1, ..., 4 cycles of deformation. The atoms are coloured according to the local crystalline environment, using Common Neighbour Analysis (CNA) [18,19]. Atoms in local face-centred cubic (fcc) structure are coloured white. Atoms in local hexagonal close-packed (hcp) structure are coloured light grey. Two adjacent layers of grey atoms are an intrinsic stacking fault, two grey layers with a single white layer in between are an extrinsic stacking fault, and a single grey layer is a twin boundary. Atoms, which are neither in fcc nor hcp structure, are shown in dark grey, these atoms are typically at grain boundaries or dislocation cores. A few isolated dark atoms are seen, this is because the thermal fluctuations interfere with the CNA algorithm used to identify the crystal structure.

As the deformation proceeds, a few intrinsic stacking faults appear as partial dislocations move through the grains. At later stages of deformation, extrinsic stacking faults and twin boundaries also appear; these are caused by partial dislocations moving on two or more adjacent slip planes. Yamakov *et al.* [20] have recently described the appearance of deformation twins in simulations of nanocrystalline aluminium, albeit with a quasi-two dimensional grain structure. As previously reported [5,6], the observed dislocation activity in the grains only account for a fraction of the plastic deformation, the majority occurs in the grain boundaries.

It is clearly seen from figure 1 that the grain structure gradually coarsens. The main coarsening mechanism seems to be that some grains grow at the expense of others, leading to a coarser grain structure with fewer, larger grains. Occasionally, low angle grain boundaries between grains with similar orientation simply vanish as the grains rotate into identical orientation. After three to four cycles, the grain size is comparable to the simulation cell, and the simulation ceases to be a realistic model of a nanocrystalline sample. As the system evolves further, the last grain boundaries disappear, leaving a single crystal behind.

To investigate the mechanism behind this coarsening, the simulations were repeated with different strain rates and temperatures. To compare the coarsening rates, figure 2 shows the number of atoms in a local environment, which is neither fcc nor hcp. This number correlates strongly with the total grain boundary area, as the far majority of such atoms are found in the grain boundaries. Figure 2a shows the number of grain-boundary atoms as a function of simulated time, figure 2b shows it as a function of the number of cycles (or, equivalently, the accumulated strain). It is clearly seen that the amount of strain required to achieve a given amount of coarsening is relatively insensitive to both the strain rate and the temperature, within the ranges investigated here. This shows that the strain is inducing the grain coarsening.

4. Discussion.

During the deformation, a large amount of mechanical work is done on the system. Most of this energy is deposited in the grain boundaries, and one could speculate that the observed coarsening is simply due to heating of the grain boundaries. However, the thermal conductivity is relatively large, and the heat is quickly spread to the entire grains (and then removed by the Langevin dynamics). During the simulation, the temperature of the grain boundaries does not rise significantly over the average temperature of the sample, so it does not seem likely that the coarsening is due to local heating. To check this assumption, a simulation was performed without any deformation. Instead, the same amount of energy was deposited directly into the grain boundaries as thermal energy. This was done by increasing the fluctuating force in the Langevin dynamics, but only for the atoms in the grain boundaries. No grain coarsening was seen in this case, providing further evidence that the coarsening is caused by the applied strain.

A sliding event consists of a few atoms moving from one position to another. Both positions are local minima of the total energy of the system. This is illustrated in figure 3, where the energy is shown as a function of a generalized reaction coordinate,

representing the collective motion of the involved atoms. As the strain is increased, some minima are favoured over others, and at sufficient strain the system moves from one minimum to another. Thermal vibrations may help overcome the last remaining barrier, although the brief timescale of molecular dynamics simulations make such thermally activated processes unlikely to happen until the barrier has become comparable to the thermal energy (kT). At lower strain rates, inaccessible to molecular dynamics, these same processes would occur earlier through thermal activation, i.e. the system would shift from state A to state B in figure 3 already when the strain has reached the level corresponding to the curve (b). This is the cause for the weak strain rate dependence of properties such as the yield and flow stresses, observed in previously published simulations of nanocrystalline metals [6]. It remains an open question if *different* processes would occur if it were possible to do the simulations with much lower strain rates. Simulations of nanocrystalline palladium provide some evidence that this is not the case, by showing that the activation energy of the grain boundary gliding is the same as for grain boundary self-diffusion [11].

The simplified view of the fundamental processes shown in figure 3 can also help understanding the observed strain-induced coarsening. In many cases the main difference between two nearby configurations, such as A and B in figure 3, is that one (or perhaps a few) atoms in a grain boundary moves from a position corresponding to a lattice position in one grain to a position corresponding to a lattice position in another grain. If this reduces the total grain boundary area, that will give an additional (small) reduction of the total energy of the system. In average, there will therefore be a tendency to favour processes that reduce the grain boundary area, leading to grain coarsening.

There does not appear to be any direct experimental observations of such a coarsening process in nanocrystalline metals, but a similar effect has been observed in nanocrystalline ceramics undergoing severe plastic deformation in a high-energy ball mill. Mørup *et al*. [21] studied the evolution of the grain size in nanocrystalline samples of α-Fe_2O_3 and ZnS, and found that while the grain size decreased in samples with a large initial grain size, it increased in samples with a sufficiently small grain size, the critical grain size being around 15 nm. Since deformation mechanisms are quite different in ceramics and metals, these experimental results cannot be taken as direct evidence for the mechanisms reported here. However, from these simulations it can be predicted that a similar effect should be seen in metals. Indirect experimental evidence is also provided by the apparent impossibility to reach grain sizes below 50 – 100 nm when nanocrystalline metals are produces by equal channel angular extrusion and similar methods involving severe plastic deformation (see e.g. ref. [4]).

5. Conclusion.

Molecular dynamics simulations have been used to investigate the behaviour of nanocrystalline copper under cyclic deformation. A major part of the deformation occurs in the grain boundaries. As a consequence thereof, the grain boundary mobility is enhanced during the deformation, leading to grain boundary migration and a coarsening of the grain structure. This effect will be dominating at small grain sizes, at much larger grain sizes severe plastic deformation is known to cause grain

refinement. It is therefore predicted that there is an intermediate grain size, towards which a system undergoing severe plastic deformation will evolve. This intermediate grain size obviously depends on the material (including the presence of grain boundary impurities), and is probably also weakly dependent on the deformation rate and temperature.

Acknowledgements.

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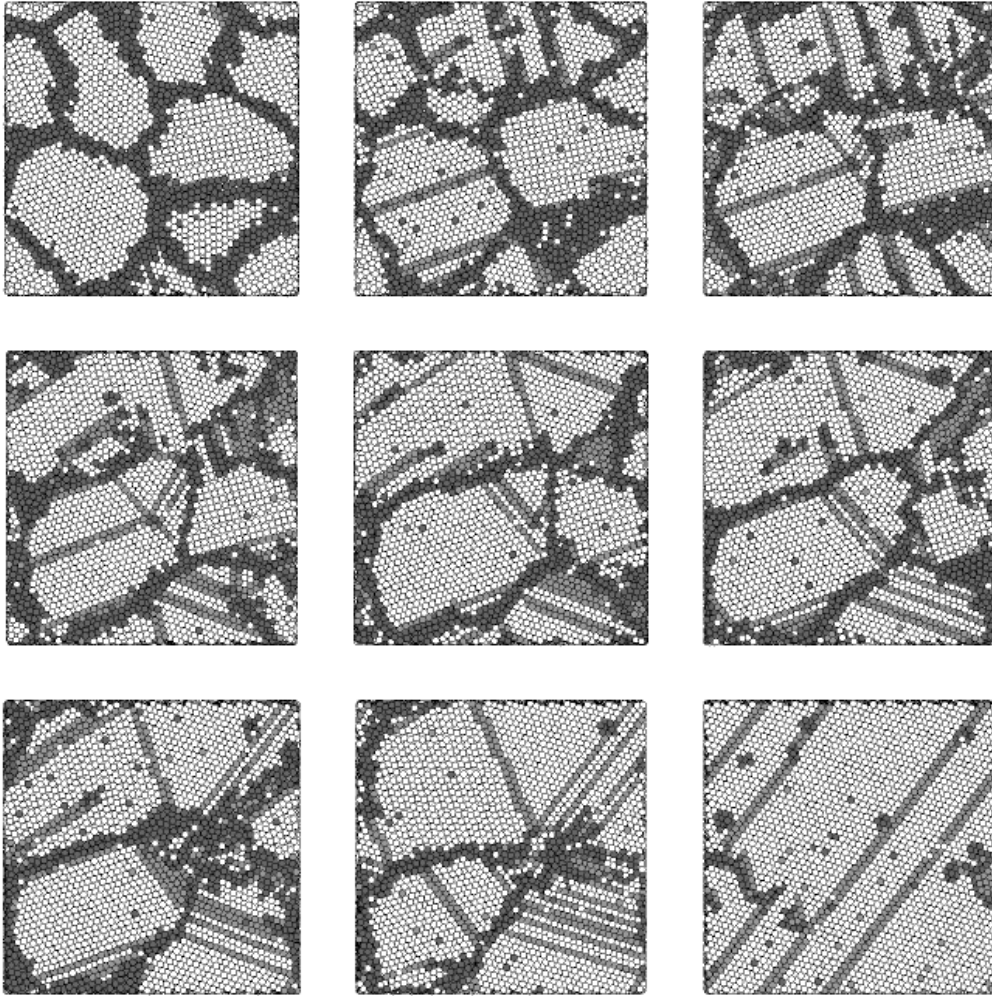21 S. Mørup, J. Z. Jiang, F. Bødker, and A. Horsewell, Europhysics Letters 56 (2001) 441.

Figure 1: The evolution of a simulated sample of nanocrystalline copper during four cycles of plastic deformation. The atoms are coloured according to the local crystalline environment. Light grey atoms are at stacking faults and twin boundaries; dark grey atoms are mostly at grain boundaries and dislocation cores. See text for details. The first figure shows the initial configuration, the second shows the system after ½ cycle of deformation, where the strain is back to 0%. The third picture is after a full cycle, etc.
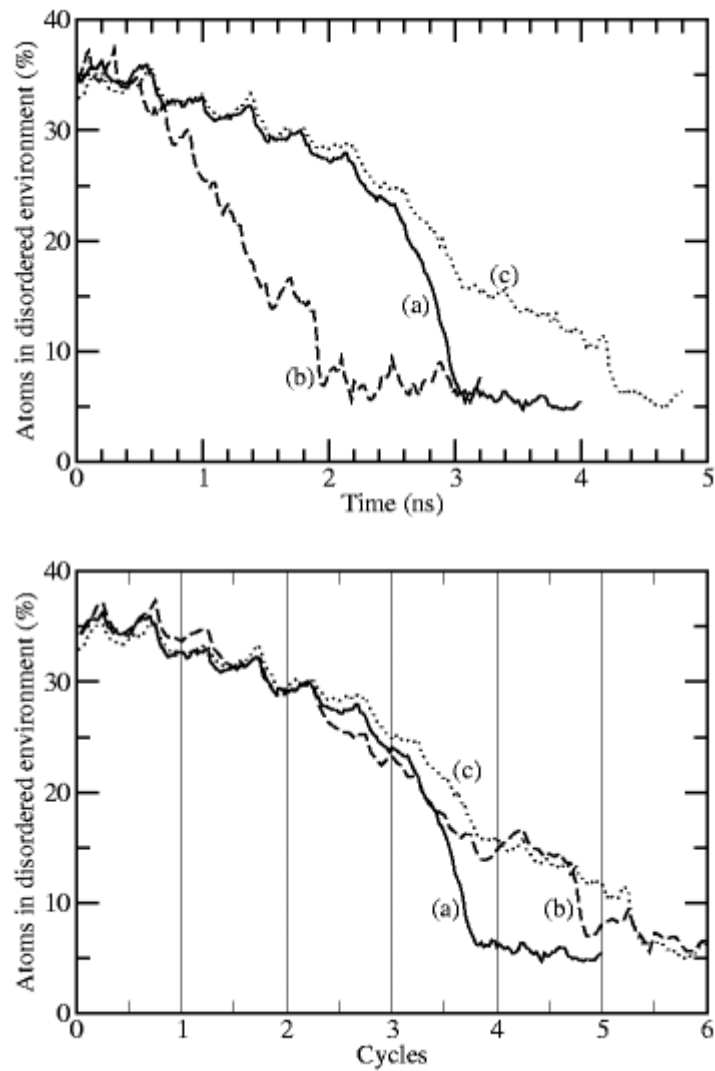
Figure 2: The fraction of atoms in a disordered environment (mainly grain boundary atoms) as a function of time (upper graph) and as a function of the number op deformation cycles (lower graph). Three simulations are shown, performed at different temperatures (T) or with different strain rates (ε'). Curve a (full line): T = 300 K, ε'= $5 \cdot 10^8$ s^{-1}. Curve b (dashed line): T = 300 K, ε'= $1 \cdot 10^9$ s^{-1}. Curve c (dotted line): T = 50K, ε'= $5 \cdot 10^8$ s^{-1}.
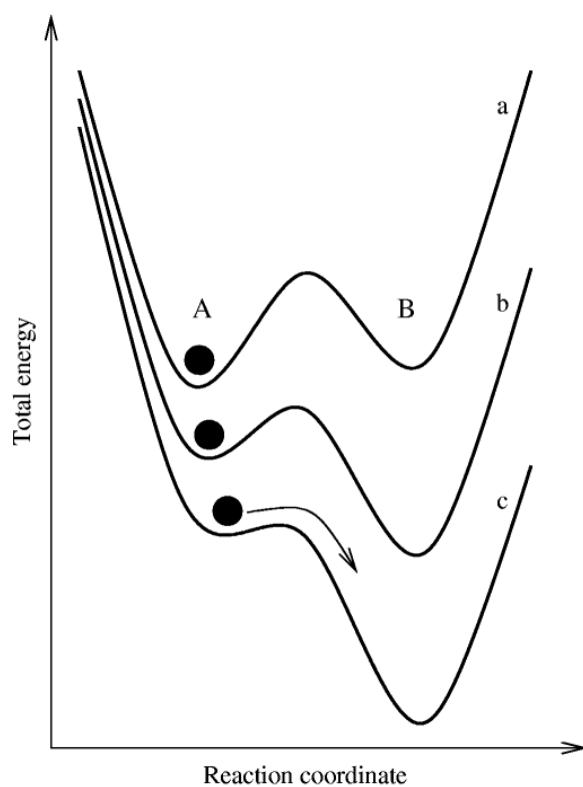
Figure 3: A sketch of the energy landscape, showing the total energy as a function of a "reaction coordinate". There are two local energy minima, marked A and B. The reaction coordinate represents a direction in the 3N-dimensional configuration space, leading from configuration A to B. The energy landscape evolves from (a) to (b) to (c) as the strain is increased; at (c) configuration A ceases to be a local energy minimum, and the system spontaneously transforms to configuration B.